Comment on "Elastic Stabilization of a Single-Domain Ferroelectric State in Nanoscale Capacitors and Tunnel Junctions"

In a recent Letter [1], Pertsev and Kohlstedt (PK) claim that "even nanoscale capacitors and tunnel junctions may have out of plane polarization sufficient for memory applications." Here we show in an elementary way that this conclusion is not substantiated by their calculations and that they should have come to the opposite conclusion within their approximations.

Indeed, while obtaining their results for the critical thickness $t_{c,\max}$ (e.g. Fig.2 [1]) Pertsev and Kohlstedt "simplified the problem by setting the gradient coefficients $g_{\ln}$ to zero". Physically, this means that the domain wall energy and the width were set to zero. Since PK consider a capacitor with imperfect metallic electrodes (or nonFE layers near perfect electrodes) there would be a depolarizing field in a homogeneously polarized film with short-circuited electrodes, and it costs *nothing* to the system to eliminate it by creating domains when $g_{\ln} = 0$. Therefore, they presented results that refer to states *unstable* with respect to domain formation. Indeed, the stability of the considered homogeneous states is governed exactly by those gradient terms that PK have nullified. When interested in a real situation, one has to use the data or the first-principles calculations to estimate $g_{\ln}$. An example of such an analysis using neutron scattering data for $BaTiO_3$ can be found in [2, 3, 4]. As follows from the discussion there, a real challenge for the material science consists in finding electrodes, ferroelectrics, and substrates to ensure a practical stability of the homogeneous polarization. PK has not even touched upon this chief problem.

PK believe that the elastic energy associated with the domain structure prevents its formation. However, this energy definitely plays no role within their approximation where the equation of state admits solutions with strictly rectangular distribution of the polarization $P(x) = \pm p$, $P^2(x) = p^2 = \text{const}$. For p equal the polarization of the homogeneous state, the strains corresponding to this solution are *exactly* the same as for the homogeneous state, i.e. the elastic energy associated with the domains is strictly zero. This does not mean, of course, that the elasticity plays no role in formation of 180° domain structure, but this role is misunderstood by PK. Hence, the criticism of prior works, Refs.[17,18] in [1], for "overlooking" the elasticity is unfair: its account is irrelevant when one defines the point of stability of the paraelectric phase with respect to the domain formation studied there. Indeed, the stability loss involves solution of a system of linear equations for the "polarization waves" while the striction term provides a nonlinear contribution renormalizing the term BP^4 in the thermodynamic potential. This term begins to play a role when the amplitude of the "polarization waves" has to be found but the correction introduced are not essential when one considers a second order transition studied there. The effects of striction in the paraelectric phase, or, more generally, effects of quadratic coupling of the order parameter with strains are indeed important when one considers loss of stability of a nonsymmetric phase with temperature or size-effect driven phase transition to the symmetrical phase. This was understood long ago for bulk transitions [5] and should be reconsidered for phase transitions in strained films, but discussion in Ref.[1] is irrelevant there.

The main point of this Comment is not that the calculation in Ref.[1] are incorrect, as they are, but that the PK approach gives no clue about the possibilities of memory in nanoscale FE capacitors. Indeed, the question they address (as prior Refs.[17,18]) is a stability with respect to very small fluctuations. Lack of such stability means, of course, impossibility of a memory. But the stability can mean two different things: an absolute stability (absolute minimum of energy) and a relative stability, i.e. metastability. In the latter case a memory is a question of time of escape from the metastable state. If this time is too short for applications, we have the case of practical absence of memory. The authors do not even mention this possibility. This is surprising given a classic example in ferroelectricity where the instability of homogeneously polarized state has little to do with polarization switching. Recall that the field at which the homogeneously polarized state becomes unstable is one or two orders of magnitude larger than the experimental coercive field. In other words, the switching occurs *not* because of instability but because of domain nucleation and growth.

Similarly, a memory loss in all likelihood is not a question of instability with respect to infinitely small fluctuations but a question of the domain nucleation, see [4]. We can also refer to the data of Noh's group [6] where the "critical thickness for ferroelectricity" and "the critical thickness for memory" are found to be quite different. The key theoretical problem to find the "critical thickness for FE memory" is that of calculating the escape time from metastable states. One has to look for adequate approaches to the problem of ferroelectric memory and, unfortunately, Ref. [1], with a true faith bestowed on numerical results without doing a qualitative analysis, does not help but rather grossly misleads this effort.

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PACS numbers: 77.80.Dj, 77.55.+f, 77.22.Ej

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